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SENSITIVITY STUDIES FOR AN IN-SITU PARTIAL DEFECT DETECTOR (PDET) IN SPENT FUEL USING MONTE CARLO TECHNIQUES

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ABSTRACT

This study presents results from Monte Carlo radiation transport calculations aimed at characterizing a novel methodology being developed to detect partial defects in Pressurized Water Reactor (PWR) spent fuel assemblies (SFAs). The methodology uses a combination of measured neutron and gamma fields inside a spent fuel assembly in an in-situ condition where no movement of the fuel assembly is required. Previous studies performed on single isolated assemblies resulted in a unique base signature that would change when some of the fuel in the assembly is replaced with dummy fuel. These studies indicate that this signature is still valid in the in-situ condition enhancing the prospect of building a practical tool, Partial Defect Detector (PDET), which can be used in the field for partial defect detection.

INTRODUCTION

Various attempts have been made in the past two decades to develop a technology to identify a possible diversion of pins and to determine whether some pins are missing or replaced with dummy or fresh fuel pins. However, to date, there are no safeguards instruments that can detect a possible pin diversion scenario that meet the requirements of the IAEA. The FORK detector system [1-2] can characterize spent fuel assemblies using operator declared data, but it is not sensitive enough to detect missing pins from SFAs. Likewise, an emission computed tomography system has been used to try to detect missing pins from a SFA [3]. This has shown some potential for identifying possible missing pins but the capability has not yet been demonstrated, especially in an inexpensive, easy to handle setting for field applications.

A novel methodology is being developed to detect partial defects in PWR spent fuel assemblies without relying on any input from the operator. An earlier paper detailed the development of a unique signature that can be obtained using a combination of neutron and gamma signals that will be noticeably perturbed if some of the fuel pins in an isolated assembly are replaced with dummy pins [4]. The objective of these simulations is to evaluate the impact on this signature when the fuel assembly is surrounded by other fuel in the spent fuel pool storage rack that typically contains dissolved boron in it.

Lifting SFAs from the storage rack and isolating them in the pool is not only an expensive operation but can also potentially cause a fuel-handling accident. As a result of these concerns there can be an unwillingness on the part of the operator to conduct such an operation. The ability to perform the measurements and obtain these signatures in an in-situ condition will represent a major breakthrough as it will be carried out in a minimally intrusive manner. The proposed instrument, PDET, is aimed at such a goal of pin diversion detection without any fuel movement and complete non-reliance on operator supplied data. It will consist of fission chambers that will principally measure the thermal neutron fields at the various guide tube locations. In addition the instrument cluster will have ion chambers for gamma detection. It is also proposed to use to the extent possible, existing hardware and equipment for performing the measurements.

COMPUTATIONAL MODEL

The base model for this study will consist of the same 14x14 PWR fuel that was used in the isolated SFA case [4]. This fuel assembly had an extreme variation in burnups ranging from 23 MWd/kg at one corner to 40 MWd/kg at the diagonally opposite corner. This SFA will be referred to as the base SFA. A

computational model consisting of a matrix of 3x3 base SFAs was developed with the assemblies in a spent fuel storage rack (see Figure 1). In the assemblies in Figure 1, the red fuel pins indicate the highest burnup with the green/blue representing the lowest burnup. The central assembly will constitute the test assembly. The other eight assemblies are oriented in a random manner around the test assembly.

An initial enrichment of 3.8 w% U-235 was selected and the varying burnup levels in the 179 active pins in the assembly were grouped into a set of nine distinct burnup levels [4]. The fuel was depleted on a single pin basis for 883 days using ORIGEN-ARP [5] to each of the nine desired burnup levels by adjusting the power in the pin. The gamma and neutron source terms and spectra and isotopics were obtained from these calculations at a cooling time of ten years. The boron concentration in the pool was 2000 ppm. The list of isotopes used was pared down to 41 (both fission products and actinides) that represented the principal absorbers [6].

The MCNP [7] calculations were performed in the fixed source mode using the latest available ENDF/B-VI data sets [8]. Two separate runs were made: a coupled neutron-gamma run using the neutron source terms and gamma run using the gamma source term. It must be noted that the neutron induced gamma contribution is negligible in these situations compared with the fission product decay gammas. Neutron and gamma fluxes were obtained at the sixteen guide tube locations (identified by the white circles in the test assembly in Figure 1) where the measurements would be made. The SFAs are in a high density storage rack with a stainless steel frame and boral (~ 4mm). For pin diversion scenarios the missing fuel pins were replaced by stainless steel rods. All relevant results had standard deviations of less than 0.5%.

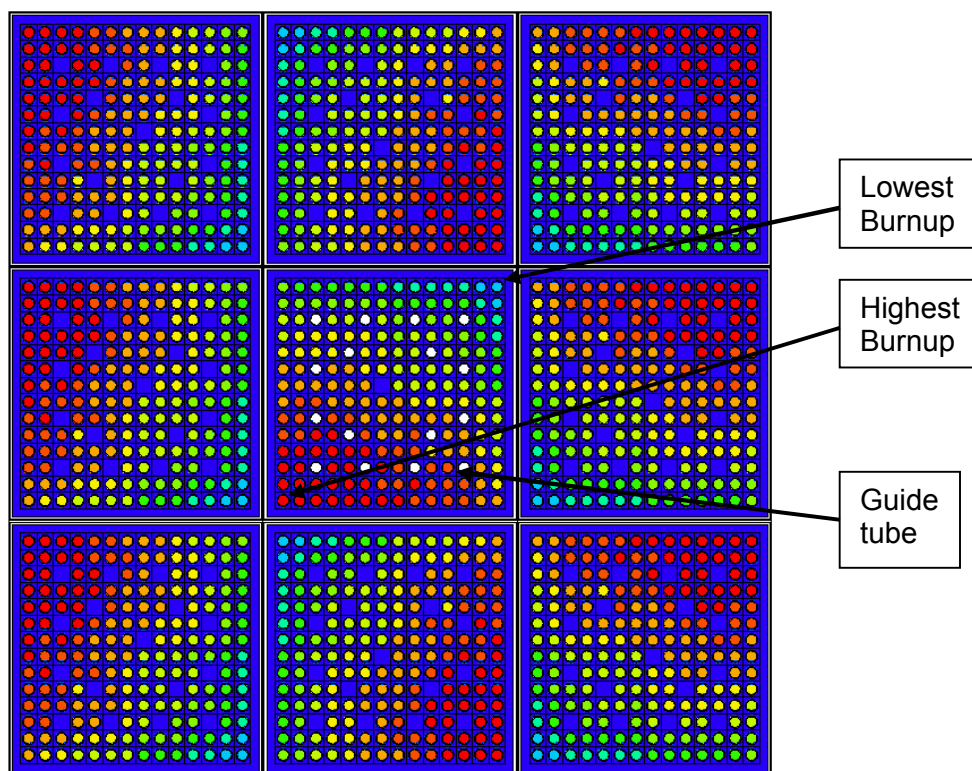


Figure 1. 3x3 Spent Fuel Assemblies in Storage Rack

ANALYSIS OF BASE CONFIGURATIONS

Since SFAs in a pool can be surrounded by assemblies of different burnups, the nine assemblies were rotated randomly to capture the impact of principally neutron leakage from the surrounding assemblies owing to differing burnups, and consequently, different leakage scenarios. There will be a much smaller amount of gammas from neighboring assemblies because of the shielding provided by the fuel pins. A set of seventeen calculations was performed to examine the impact of the surrounding assemblies on the neutron and gamma signals.

The base signature developed is the ratio of the gamma to thermal neutron signal at each of the sixteen guide tube locations normalized to the maximum among them. Figure 2 shows the alphabetic labels 'a' through 'p' for the sixteen locations. The base signature is developed by plotting the normalized ratios from each of the four clusters of guide tubes starting at the center location and going counter clockwise around the cluster of four locations (e.g. c, d, a, b, etc.)

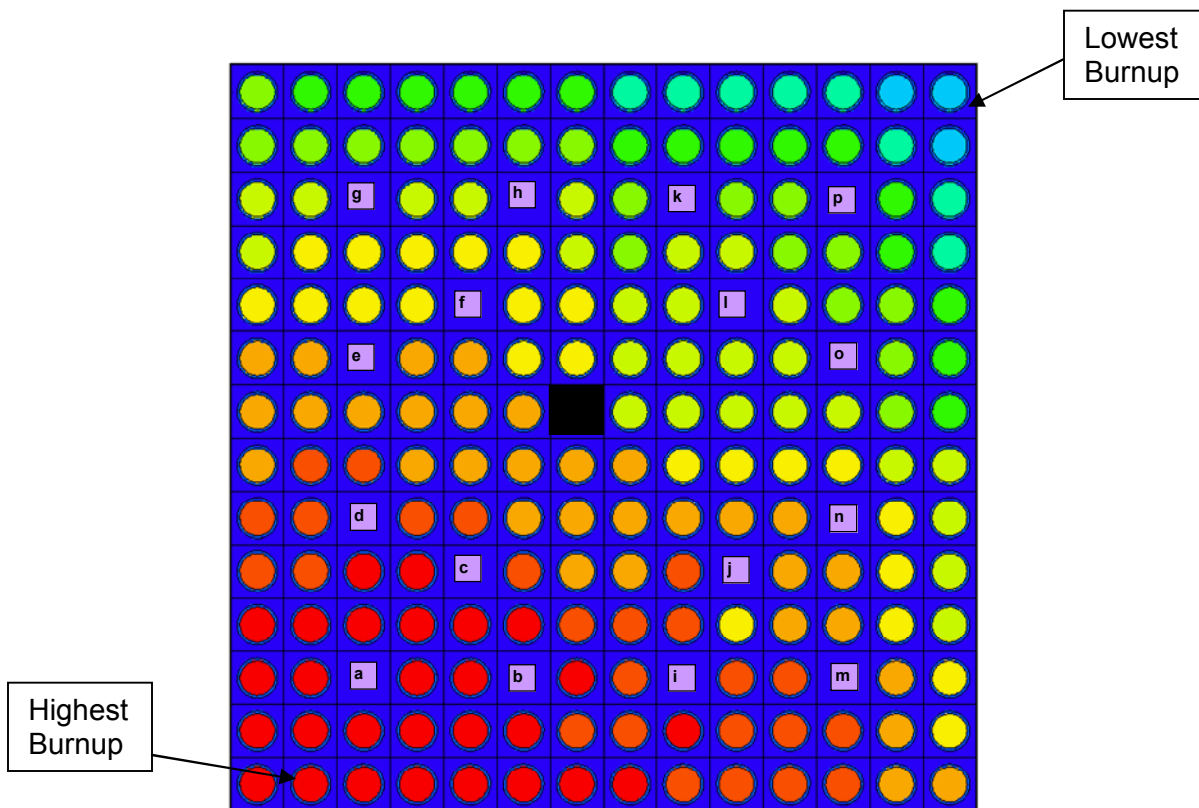


Figure 2. Spent Fuel Assembly with Guide Tube Labels

Figure 3 shows the mean distribution from the seventeen random rotations with the associated uncertainty. The largest uncertainty is approximately 0.075 at the 1-sigma level. The signature shows a tilt towards the low burn-up region of the assembly (locations k, l, o, and p in Figure 2). The gamma signal is roughly directly proportional to the burnup. It is a more localized signal because of the heavy attenuation by the fuel pins themselves. Neutron responses are very strongly dependent on burnup, usually varying as the 3rd to as much as the 5th power of the burnup at enrichments present in PWRs. The neutron signals are a result of both spontaneous fissions from transuranics as well as induced fission in the fissile material in the pins [9]. These fission neutrons migrate and slow down to thermal energies resulting in a less localized signal that is caused by particles from more than just neighboring pins. In an isolated assembly, the corner locations see less of a thermal neutron signal than do the interior locations

since these have contributions from fewer pins than do the interior locations. However, when assemblies are surrounded by other assemblies, there will also be neutrons migrating between assemblies. The net leakage will be towards regions with lower neutron populations such as the low burnup regions. The fission neutrons are only minimally impacted by the presence of boron in the system. Once thermalized, they contribute to the neutron signal in the local guide tube locations. While there is some contribution to the gamma signal from neighboring assemblies this is minimal owing to the presence of the high-density, high-Z material in the form of fuel.

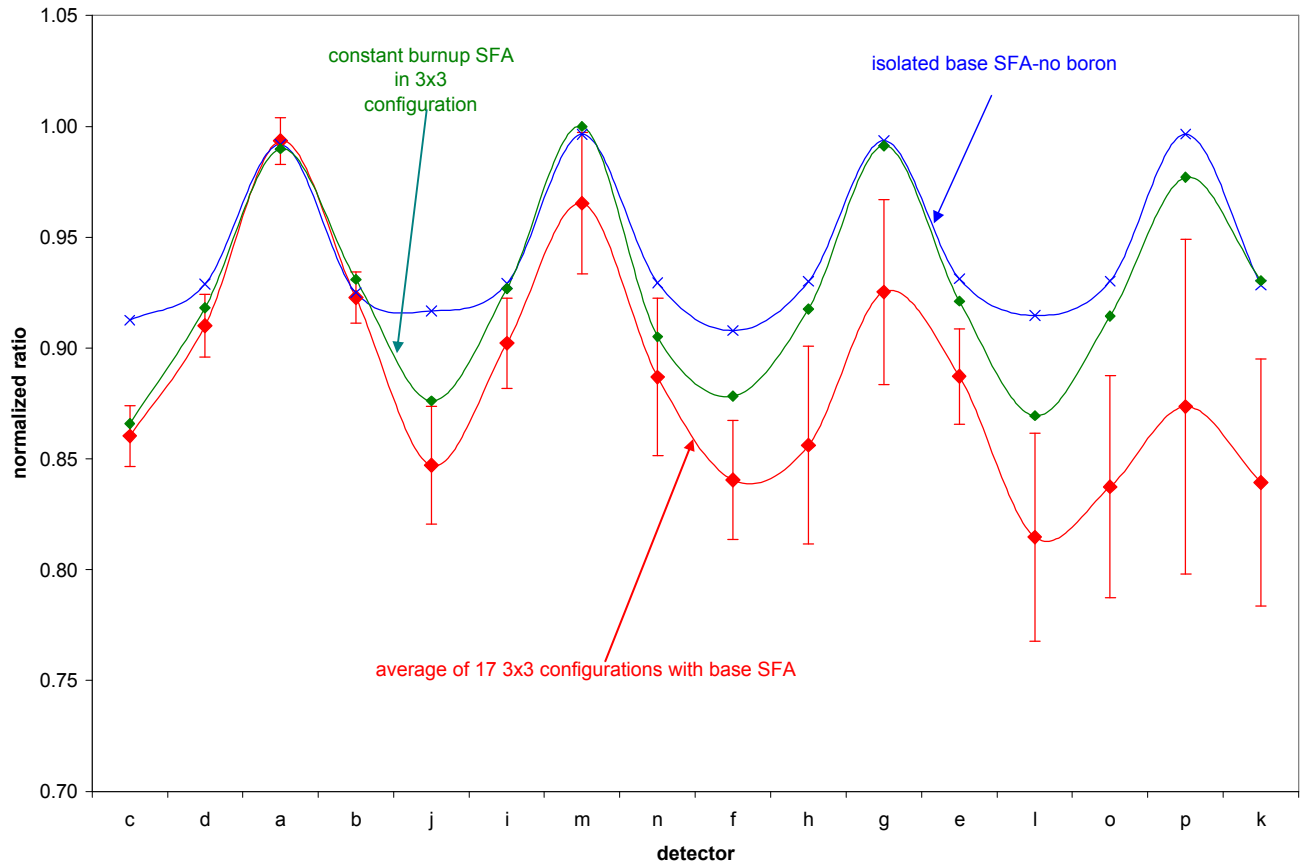


Figure 3. Signature from Seventeen Random Orientations of the 3x3 Configuration

Therefore, while the gamma signal is less sensitive to the presence of neighboring assemblies, the thermal neutron signal increases, with this increase being greater in the lower burnup regions of an SFA. This accounts for the tilt of the signature seen in Figure 3. It is also noted that the uncertainty in the low burnup region is greater since the random rotations can place high as well as low burnup pins in the neighborhood of the low burnup part of the central assembly. Figure 3 also presents the signature from a case where all nine SFAs have constant burnups as well as the signature from the base SFA isolated with no dissolved boron. When all SFAs have the same burnup the net migration from neighboring SFAs is minimal and the tilt in the signature virtually disappears and is very close to the signature from an isolated SFA with no dissolved boron.

In addition to the impact of neighboring SFAs, the impact of dissolved boron on the signature was also studied. Figure 4 presents the signatures for four cases of a single 3x3 configuration with the base SFA: 0, 1000 ppm, 1500 ppm, and 2000 ppm.

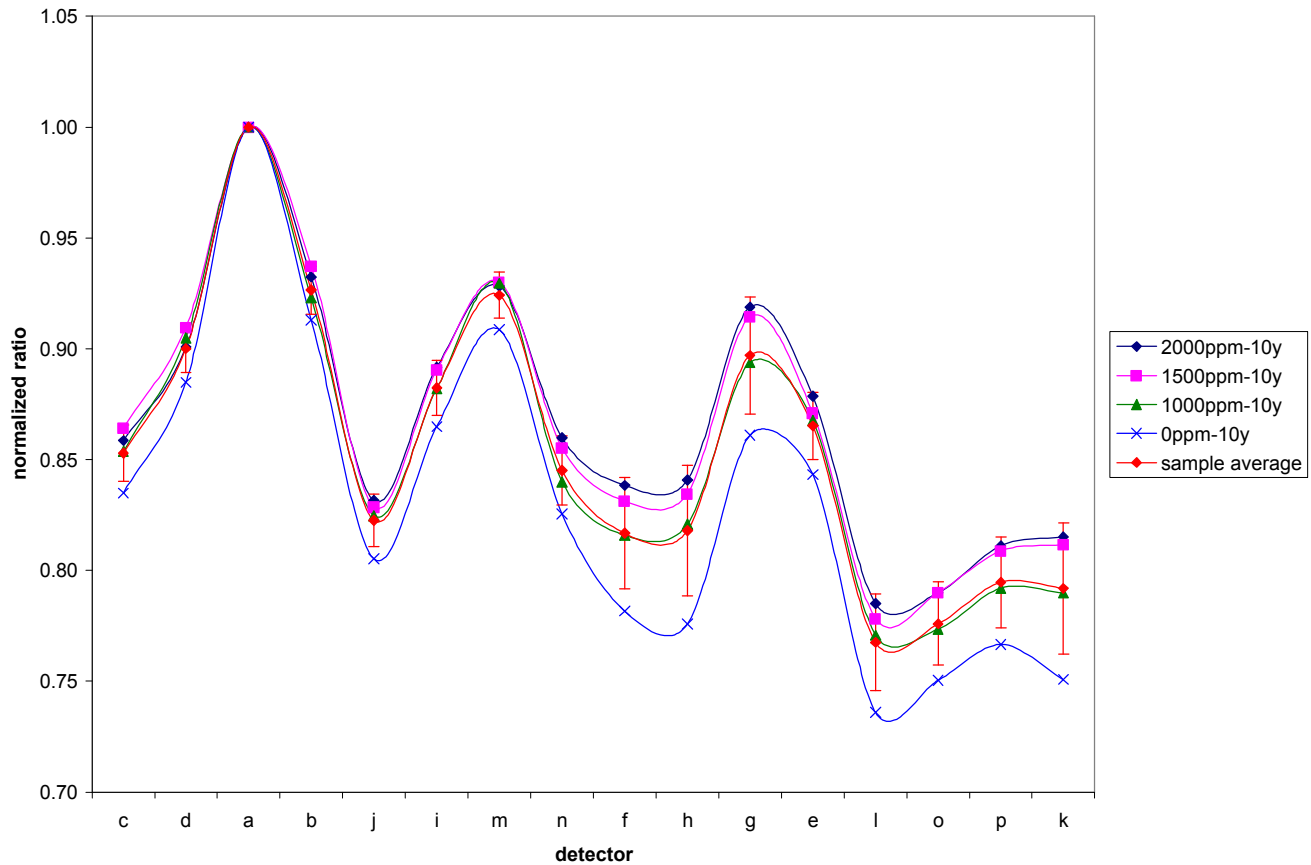


Figure 4. Sensitivity of Signature to Boron Concentration

The boron content in the pool tends to flatten the signature, as is evident from Figure 4. The shorter mean free path of the thermal neutrons renders the thermal neutron signal to become more localized thus reducing the thermal neutron population at the guide tubes. The mean of the four cases shows a similar trend as that of the seventeen random rotations (see Figure 3) though with a smaller uncertainty, the largest being approximately 0.03.

Sensitivity to both initial enrichment of the fuel and cooling time were also studied. Typically, the neutron signal is affected by both these parameters. As in the case of boron, the gamma signal remains mainly unaffected by the initial enrichment and cooling time. This is because the principal gamma signal comes from the long lived ^{137}Cs which has similar yields for both ^{235}U and ^{239}Pu . Thus the impact of these parameters on the signature is due to the relative change in the neutron signal among the various guide tube locations. Studies with single SFA cases indicate that this effect is of the order of about 0.02 each on the signature.

A base signature can therefore be established that retains a shape that is principally influenced by the location of the guide tube in the SFA. As explained earlier, the corner locations which are less influenced by surrounding fuel pins form the peaks of this signature while the central locations form the low points, i.e., the neutron and gamma signals are higher at the central locations. The central to corner guide tube gamma signal ratio is larger than that of the thermal neutrons. The other locations make up the rest of the shape transitioning from the peak to the valley in the signature. The presence of surrounding SFAs in an in-situ condition tilts the signature downward in the low burnup regions because of the relatively higher neutron migration from these SFAs to these regions compared with the migration

to the high burnup regions. The amount of downward tilt towards regions of lower burnup will vary depending on the burnup in the regions of the neighboring SFAs that border these regions. The base case SFA examined here has a large burnup gradient; more uniform burnup cases tend to make the signature more symmetric (see Figure 3). Gamma signals are almost entirely local and little leakage occurs from adjacent assemblies.

It must be borne in mind that this signature can be such that any of the locations a, g, m, or p can have the highest relative ratio of 1 and the location c, f, j or l can have the lowest relative ratio. Thus, it would be useful to construct bounding shapes based on averages of each of these sets as well as the remaining eight guide tube locations and applying uncertainties on them. The constraint on this will be that the corner locations can have a maximum value of 1. The uncertainty has been calculated as the square root of the sum of squares of the maximum uncertainty from each of the sensitivity studies described above. This value rounded up is 10%. Applying this uncertainty, a maximum and minimum bound of the average base signature has been constructed. Deviation of shape and magnitude that beyond these bounds would indicate missing fuel from a SFA. Figure 5 shows these base curves.

PIN DIVERSION CASES

Pin diversion cases where 22 pins out of 179 were replaced with stainless steel rods were studied for two different 3x3 configurations. This represents about 12% of the pins missing from the SFA at the center of the 3x3 configuration. Figure 5 shows the deviation in two of these cases.

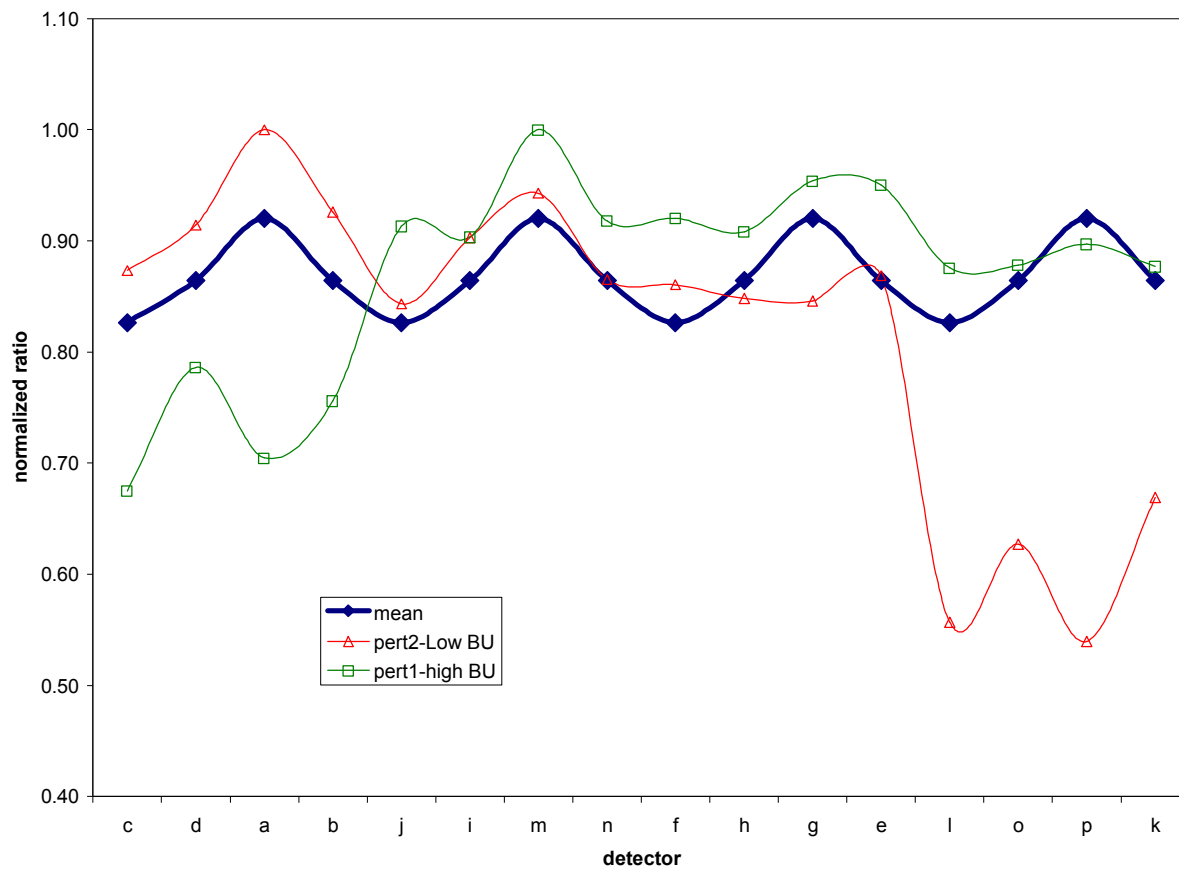


Figure 5. Base Signature with Two Pin Diversion Scenarios

In the first case the bulk of the missing pins were in the vicinity of the 23 MWd/kg guide tube locations - 2 in the vicinity of 'k', 7 in the vicinity of 'p', 3 in the vicinity of 'o', 5 in the vicinity of 'l', and 2 further out in the vicinity of 'g'. It is clear from Figure 5 that the signature has changed and the affected parts of the signature are outside the bounds of the uncertainty of 10% in the shape. Even location 'g' with only two missing pins shows a deviation in shape.

The second case had a perturbation with the bulk of the pins missing around the high burnup region of 40 MWd/kg - 6 in the vicinity of 'a', 2 in the vicinity of 'b', and 4 in the vicinity of 'b', 2 in the vicinity of 'd', and the rest further away. The shape has again changed though to a lesser extent than when the bulk pins were missing in the low burnup region.

The gamma signal drops in the vicinity of the missing pins mainly due to the loss of the local source of gammas. The drop in the gamma signal contributes to the drop in the relative ratio to a large extent. There is an increase in the thermal neutron population at the locations in the low burnup region in the vicinity of missing pins because of the migration of neutrons from the high burnup regions that are intact as well as the lack of fuel that absorbs the thermal neutrons. For the case of missing pins in the high burnup region, there is a slight increase in the locations surrounded by a few missing pins (e.g., location 'a' with six pins missing) for reasons just discussed. The impact on the neutron population is smaller in the high burnup regions with missing pins unless a very large number of the high burnup pins is missing. The deviation of the perturbed signature from the base signature is attributable in part to the drop in the gamma signal and an increase in the neutron signal in the vicinity of the missing pins. The surrounding SFAs have a larger influence on the change in the magnitude of the neutron signal than they do on the more localized gamma signal.

A combination of the drop in the gamma signal combined with increases in the thermal neutron signal makes the relative ratio in the signature drop, leading to an overall change of shape in the signature that can be visually detected.

CONCLUSIONS

The results from the studies presented here indicate that partial defects in PWR SFAs can be detected in an in-situ condition. The base signature from an intact SFA located in the storage rack maintains the same basic shape, principally geometry dependent, which it did for an isolated SFA. The deviation from the base signature both in magnitude and shape can be ascertained by visual inspection. The proposed instrument, PDET, which can perform these measurements, has the potential of being a powerful and practical tool for use in the field in a minimally intrusive manner and without relying on any operator provided data on the SFAs. Unique signatures for other PWR fuel types such as 17x17, 15x15 etc. can be similarly developed.

A set of controlled experiments is planned to test and validate this novel methodology. These tests will involve real PWR SFAs, some of which have missing fuel. They will aid in not only validating the predictions of the simulation studies but also provide data to potentially refine the methodology that promises to be a breakthrough in partial defect detection technology.

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REFERENCES

- [1] Titta, et al, "Investigation on the possibility to use FORK detector for partial defect verification of spent LWR fuel assemblies," Final report on Task JNT A 1071 of the Member State's Support Programme to IAEA Safeguards, 2002.
- [2] B. D. Murphy and P. De Baere, "Monte Carlo Modeling of a Fork Detector System," 27th Annual Meeting, Symposium on Safeguards and Nuclear Material Management, London, England, May 10-12, 2005.
- [3] F. Levai, et al, "Feasibility of gamma emission tomography for partial defect verification of spent LWR fuel assemblies," Task JNT 1201 of the Finland, Hungary and Sweden to the IAEA safeguards, 2002.
- [4] S. Sitaraman and Y.S. Ham, "Characterization of a Safeguards Verification Methodology to Detect Pin Diversion from Pressurized Water Reactor (PWR) Spent Fuel Assemblies using Monte Carlo Techniques," 48th Annual Meeting of the Institute of Nuclear Materials Management, Tucson, Arizona, July 2007.
- [5] ORIGEN-ARP, Version 5.1.01, Isotope Generation and Depletion Code, CCC-732, Radiation Safety Information Computational Center, March 2007.
- [6] S.P.Cerne, O.W.Hermann, R.M. Westfall, "Reactivity and Isotopic Composition of Spent PWR Fuel as a Function of Initial Enrichment, Burnup, and Cooling Time", ORNL/CSD/TM-244, Oakridge National Laboratory, October 1987.
- [7] X5-Monte Carlo Team, "MCNP-A General Monte Carlo N-Particle Transport Code," Version 5.1.40, Los Alamos National Laboratory, February 2006.
- [8] Evaluated Nuclear Data Files, National Nuclear Data Center, Brookhaven National Laboratory, various dates.
- [9] J.R.Phillips, "Irradiated Fuel Measurements", in "Passive Nondestructive Assay of Nuclear Materials", NUREG/CR-5550, edited by D. Reilly et al., March 1991.